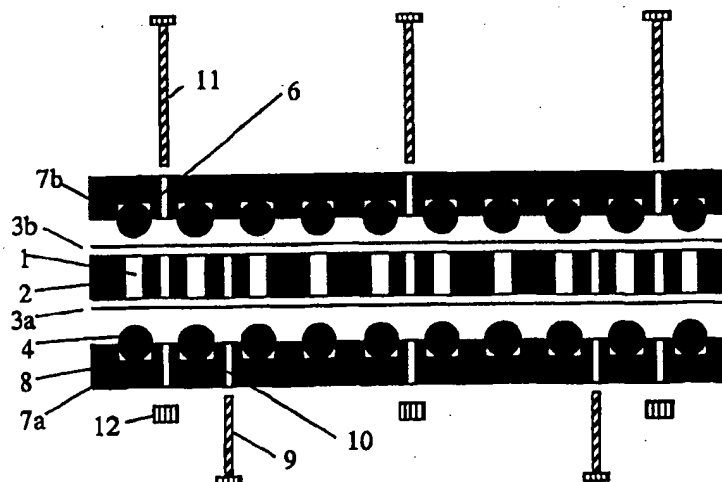




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(54) Title: MULTIAUTOCCLAVE FOR COMBINATORIAL SYNTHESIS OF ZEOLITES AND OTHER MATERIALS

**(57) Abstract**

The invention relates to a multiautoclave and details of its design and a method for automated synthesis of zeolites in said multiautoclave and, furthermore, application of the multiautoclave for an automated synthesis which is optimized simultaneously with regard to several synthesis parameters in the synthesis of zeolites. The multiautoclave consists typically of a pressure vessel/autoclave lined with an inert material with from 10 to 10.000 small, separated chambers, each typically having a volume of 0,2-2 ml, the chambers preferably being formed as through-going perforations in a central block and the perforations are sealed by means of balls, septa, stoppers or such which are placed at the bottom and top of each through-going perforation, and metal plates are placed over and under the Teflon plate so that the closing mechanism is pressed against the edges of the perforations in the Teflon plate with sufficient load to enable the chambers to be filled with aqueous mixtures and to be heated to 200 °C without the occurrence of leakage. Top-and bottom plates and closing mechanisms can be integrated so that all the perforations are sealed simultaneously when these are placed at the top and bottom sides of the central block.

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MULTIAUTOCLAVE FOR COMBINATORIAL SYNTHESIS OF ZEOLITES AND OTHER MATERIALS

FIELD OF THE INVENTION

5 The present invention relates to a pressure and temperature reactor vessel, especially a multiautoclave and to details concerning the design of this equipment.

BACKGROUND OF THE INVENTION

Many materials, such as e.g. zeolites, are prepared by so-called hydrothermal synthesis at
10 temperatures ranging from 100°C to 200°C requiring crystallization times of 1 hour or more. For syntheses being carried out at temperatures that are higher than the solvent's boiling point, it is necessary to use pressure vessels, and these have to be suitable for the temperature and pressure used during the operation. The pressure vessel has to be designed so that the handling of it does not represent any unnecessary hazard, provided it
15 is used according to working instructions.

Zeolite syntheses are usually performed in strongly alkaline media, often at $\text{pH} > 14$, and the reaction mixture will often contain toxic chemicals, such as e.g. fluoride. Conventionally, syntheses that may be performed at temperatures lower than 110°C are carried out in polymer bottles, often Teflon, while reactions at higher temperatures require
20 steel autoclaves, perhaps lined with Teflon. The price of an autoclave of this type with the required safety details are typically of the order of NOK 10.000,- or higher. Furthermore, such an autoclave will weigh from 1 kilogram and upwards, and all these elements represent limitations regarding the number of syntheses that may be performed in most laboratories in the course of one year.

25 Zeolite synthesis is often carried out by keeping the synthesis mixture at around 100°C for at least 6 h. At these moderate temperatures sealed chambers are necessary in order to avoid drying out of the synthesis mixture.

30 As an example of conventional zeolite synthesis, Zeolite Y can be prepared according to US 3.130.007, Example 1, by dissolving 5 g sodium aluminate containing 30 weight percent Na_2O and 44 weight percent Al_2O_3 and 22 g sodium hydroxide containing 77.5 weight percent Na_2O in 89,5 ml distilled water. This solution was added to 124.2 g of an aqueous colloidal silica sol with 29.5 weight percent SiO_2 , so that the resulting mixture
35 had a composition corresponding to $13.9 \text{ Na}_2\text{O} : \text{Al}_2\text{O}_3 : 28.2 \text{ SiO}_2 : 471 \text{ H}_2\text{O}$, and the mixture was homogenized by stirring. The mixture was enclosed in a sealed glass vessel, placed in a water bath and heated at 100°C for 21 hours, after which the product was recovered by filtering, washed and dried. Common to all the synthesis procedures

mentioned and for all other known synthesis procedures for preparation of zeolites on laboratory scale with the purpose of discovering new zeolites or to optimize existing zeolites, is that these are performed in a cumbersome and expensive manner by having to separately prepare each reaction mixture, which typically consists of 4-7 reagents, and by adding the reagents one by one.

In many other examples the synthesis of zeolites and other molsieves needs temperatures well above 100°C, so that steel pressure vessels or the like are required.

Furthermore, each reaction mixture is typically prepared in batches of 5 to 100 g and crystallized in expensive and heavy autoclaves with internal volumes often in the range of 25 to 250 ml and with weights of up to 8 kg per autoclave, causing considerable expense due to a large consumption of often expensive reagents and due to the fact that the handling of the heavy autoclaves often makes it impossible to handle more than one autoclave at the time, and finally that the size of the autoclaves limits the number of autoclaves that may be placed in each oven or heating unit. The combination of all these elements are, according to known technology, making each zeolite synthesis a very resource intensive process, and there is a great need for greater efficiency, rationalization, downscaling and automation. Simple calculations have shown that by combining the different variables which are involved in zeolite synthesis with narrow enough intervals in reagent concentrations, temperatures, reaction time, etc. to cover any phase formation based on known examples, it is feasible to make up 10^{18} recipes. With to-day's synthesis capacity, which on a global basis hardly exceeds 100.000 syntheses per annum, it would take 10.000.000.000.000 years to carry out all these syntheses in which each and every one in theory has potential for the preparation of a new zeolite or other microporous material. The expenses involved in performing these syntheses according to known technology would obviously be formidable, and there is thus a great need for new and more cost efficient methods for zeolite synthesis.

In recent years new, automated methods for systematic preparation of new compounds, so-called "combinatorial techniques", have been developed, but equipment which may be used for liquid phase synthesis at temperatures above approx. 100°C has till now not been disclosed, because this requires that the synthesis takes place in a hermetically sealed vessel at elevated pressures. WO 95/12608-A1 for instance, discloses an apparatus and a method for a) synthesis of several molecules on substrates, comprising distribution of the substrates in the reaction chambers, b) combination of the first addition of these molecules with different reagents in each of the reaction chambers, c) moving the substrates through tubing to separate mixing chambers where the substrates are mixed, d)

redistribution of the substrates by transport through tubing back to the reaction chambers, and e) combination of a second portion of different composition to the first portions of molecules in the different reaction chambers in order to prepare new mixtures. This publication describes only a system for mixing and distribution of different molecules and not a system for hermetical sealing of the reaction chambers which would make it possible to operate at high temperatures, and this system would thus not be suitable for the synthesis of zeolites. In WO 96/11878 there is a description of extensive use of a combinatorial arrangement for synthesis of new materials, including zeolite synthesis at 100°C. Even though this patent application presents a detailed description of instrumentation and equipment developed for different purposes, autoclave systems required for performing the syntheses under the prevailing physical conditions (elevated pressure and temperatures exceeding 100°C) are not described.

Prior art teaches autoclaves with several chambers for special purposes, and there is for instance in US 5.505.916 a description of a metal cassette which can be opened and closed like a suitcase, and which has an interior with compartments intended for placement of the different instruments used by dentists, where these may be sterilized by autoclaving. Furthermore, large autoclaves intended for instance for the growth of crystals, are known, examples are described in US 5.322.591, US 5.312.506 and US 5.476.635, but the purpose of these and similar autoclaves is to make it possible to carry out large-scale syntheses, for which there is a great need when a synthetic procedure has been established and scale-up is desired, or when the purpose is to grow single crystals as large as possible. The autoclave described in the earlier mentioned US 5.312.506 is designed to withstand temperatures up to 1500°C for growth of crystals from metal melts. Another feature in connection with work with autoclaves is energy savings, and this is addressed in EP 0.434.890 A1, with description of a system for insulation of the autoclave walls and for the design of such insulating layers in the walls, which could be useful for large-scale autoclaving, but is of no relevance when working with small laboratory autoclaves which are heated in ovens.

Furthermore, there is a series of known equipment intended for synthesis of proteins and biopolymers, where the design comprises sheets with a large number of chambers intended for screening of syntheses and crystal growth, in its simplest form as described in US 5.096.676. US 5.400.741 describes a diffusion cell for growth of the largest and the most perfect crystals possible of macromolecular compounds by a technique called the "hanging drop" technique. Several patents, e.g. US 5.013.531, US 5.531.185, US 5.362.325 and EPA 0.553.539 A1, deal with cells for growth of proteins and biopolymer crystals in spacecrafts. Common for the latter patents is that the designs described are

very sophisticated and thus very expensive, because they are intended for use in spacecrafts. Common for all equipment designed for synthesis and crystal growth of proteins and biopolymers is that they are meant for use at low temperatures, or typically temperatures in the range of 0°C to 65°C, and that they consequently are not designed to withstand conditions typical for hydrothermal synthesis. In addition, many of these prior art synthesis cells are not lined with Teflon or other similarly inert materials, something that almost without exceptions is required for synthesis of zeolites and the like. There is, e.g., a known design called "multiblock" (Krchnak, V., Vagner, J.; Peptide Res. 3, 182 (1990)) consisting of i) a Teflon block holding 42 reactors, polypropylene syringes equipped with polymer filters, ii) a vacuum adapter connecting each reactor to a vacuum line (not described in detail) which enables rapid washing in an apparatus for continuous flow, iii) two Teflon plates with 42 stoppers to which the Teflon block is fastened during use, and iv) a glass cover used during homogenization. The problem with this design is that the reactors which are made of glass and which do not have protected sidewalls may be used only at low pressures and not in strongly alkaline solutions. There is thus no available literature describing equipment that might be used for practical work with combinatorial zeolite synthesis, in as much as such syntheses almost without exception require hydrothermal treatment of a solution or gel with relatively high content of water and often high contents of organic compounds in a closed chamber, and almost all methods for preparation of zeolites known so far require such conditions during synthesis, and this is true without exception for all methods which have proved to be commercially applicable. The synthesis of zeolites is thus normally performed under hydrothermal conditions which require elevated pressures and high temperatures in periods up to several weeks without leakage. The problem has so far been the costs involved in this type of work, estimated to an average of NOK 5.000,- per synthesis, including recovery of the product and XRD analysis. An important feature when dealing with large series of syntheses is therefore how the product can be recovered and washed in a rational way without insurmountable expense, something that is not disclosed in prior art. As far as known, this type of work is performed in the same manner by all synthesis laboratories engaged in synthesis of zeolites and non-carbon-based molecular sieves.

One objective of the present invention has been to develop a complete system for screening of synthesis conditions for preparation of zeolites and other non-carbon materials requiring hydrothermal conditions in the temperature range 100°C to 250°C in a more cost efficient manner, and it has thus been of interest to improve a series of parameters, which means making them more cost efficient. Some of these parameters are:

1. Reduced size of the separate reaction chambers and increased number of reaction chambers what is called a multiautoclave.

This will lead to reduced use of reactants and thus cheaper synthesis.

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2. Automated addition of reactants, for instance by having 100 reaction chambers present in one multiautoclave and by enabling this to be connected to a pipetting machine which makes quick and exact addition of all liquid reactants possible.
- 10 3. Simple and easy-to-use mechanism for the closing and opening of the multiautoclave.
4. Simple recovery and washing of the synthesis product and simple cleaning of the multiautoclave after use.
- 15 5. Devices allowing automated analysis with X-ray diffraction and automatic identification of known crystalline phases by combination of an automatic sample switcher, a structure library stored in a database and software that can monitor sample switching and identification.

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Another objective of the present invention described here has been to design automated equipment for larger synthesis series and prepare formulations based on mixtures of different liquids/solutions with varying reactant ratios.

- 25 These and other objectives are attained by the present invention, which represents a breakthrough in terms of cost reduction for e.g. zeolite synthesis in that the reaction mixture crystallizes in a volume reduced typically to 1/100 of what has been used conventional, thereby achieving reduced consumption of reactants and cheaper syntheses, and further by enabling automated addition of reactants, e.g. by having 100 or more available reaction
- 30 chambers in one single multiautoclave, and that the multiautoclave plates can be connected to a pipetting machine that makes quick and exact addition of all liquid reactants possible, and by being able to place several such plates with reaction chambers on top of each other without difficulty. Furthermore, an important feature of the present invention is the simple and not very time-consuming operation of the multiautoclave.

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The present invention relates to a pressure and temperature reactor vessel comprising

- a) a central block having a multitude of perforations, wherein said perforations are through-going perforations, or cavities or other form of holes permanently closed at one end,
- b) cover means, operatively associated with a sealing means, for engagement with said central block to seal the open ends of said perforations forming a multitude of chambers,
- c) a sealing means, operatively associated with the covers means, to form a pressure tight seal when said cover means is brought into position by a locking means,
- d) a locking means acting in concert with the cover means to engage the sealing means so as to define a multitude of reaction chambers.

Applications for the present invention may, in addition to zeolite synthesis, be in any field of activities within research and development connected to products where at least one production-step comprises the mixing of different liquids, e.g. in the fields of organic and inorganic syntheses, paint production, formulation of fuels, food industry, etc., and, furthermore, applications within clinical testing, dissolution and digestion of samples with acid etc. where a liquid reactant is added to a liquid or solid. The invention is in particular aimed at applications where open vessels cannot be used, and more specifically for applications where it is required to operate at temperatures which will cause elevated pressures in the liquid part of the mixture.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Further advantages and characteristic features of the present invention are illustrated in the following description by referring to the drawings which, as mere examples without limitations, show some designs related to this invention, where Figure 1 present a side-view of the multiautoclave with the single components disassembled, and Figure 2 shows a top-view of the multiautoclave, while Figure 3 shows an alternative design of the multiautoclave (side-view). Figure 4 shows a section of one of the chambers of the multiautoclave (side view) equipped with a Teflon liner and disc-shaped lids. Figure 5a shows a section of the multiautoclave in "exploded" view, and in Figure 5b in closed mode.

Figure 6 presents an example of different chemical compositions covered in one single experiment using the multiautoclave.

More specifically, the invention comprises a unit containing a multitude of pressure vessels, also referred to as a multiautoclave. This multiautoclave has typically from 10 to 10.000 or more small, separate chambers (1), each typically with a volume of 0.001 – 10 ml. The multiautoclave is composed of a set of plates (2 and 7) and optionally thin laminae (3) which are stacked so that they form a matrix of small chambers (1), as shown

in Figures 1 - 5. The thickness of the different plates and laminae may vary, always keeping in mind that a maximum number of small chambers with optimal size is desired, and that the chambers must be leak tight and dimensioned such that no excessive degree of deformation will take place under the operational conditions which the equipment is designed for. The central block consisting in a perforated plate (2), with a multitude of perforations, serve – when sandwiched between the plates (7a and 7b) and closed – as the frame of the pressure chambers, and may for use at higher temperatures (150 - 250°C) be made from stainless steel, aluminium, titanium or other rigid material s.a. PEEK or the like, with perforations of e.g. 10 mm diameters, where each perforation is lined with a segment of Teflon tubing or tubing made from another suitable polymer material, and where the walls of the linings e.g. can have a thickness of 1-3 mm. For use at temperatures below 150°C, the central block can be made entirely of Teflon, and for use below 130°C it can be made of polypropylene, and for use below 105°C it can be made of polyethylene. In the separate pressure chambers, balls (4) made from Teflon or other suitable material s.a. steel, PEEK, Nylon or glass may be used as a bottom- and toplid, also referred to as sealing means or septa (5) made from an elastomer or from another appropriate material s.a. Teflon in the shape of circular disks which will give the advantage in the form of reduced weight and volume. Septa should be made from an elastomer, preferably Viton, or other suitable material that can withstand temperatures of at least 200°C, and they may be equipped with Teflon lining (13) on the side facing the chamber as shown in Figures 4 and 5. Furthermore, said septa (5) should have a thickness considerably larger than the depth of immersion, as shown on Figure 3, in order to ensure a tight fit when assembling the different elements. As an alternative, septa or balls may be fastened on the bottom and top plates or on separate polymer films or a thin metal plate in such a way that they may be put in place and removed in a simple manner. The bottom and top plates (7) thus hold balls, septa, stoppers or other types of suitable cover means in place as the central block (2) is squeezed between the bottom and top plates. In order to avoid the use of balls, septa, stoppers or loose parts, the bottom and top plates can also be designed with a structure that has conical or hemispheric protrusions that fit into the perforations. Another design comprises polymer films moulded or otherwise shaped to cover all the perforations. In yet another design, the central block has been machined so that sharp edges (14) protrude around each perforation or well, as shown in Figure 5. The advantage of this design is that one smooth polymer sheet can be used as a lid or sealing device for all the chambers because the sharp protrusions cut into this polymer sheet so that leakage between adjacent chambers does not occur when the reaction vessel is closed with its locking mechanism. Said sharp protrusions (14) could alternatively consist of rings or e.g. a rectangular grid that are either welded onto the central block (2) or fastened by any other suitable method. These said protrusions may optionally be part of the insides

of the top (7b)- and bottom plates (7a) or the central block (2). The important point here is that the load used when assembling the different elements is concentrated just at the edges of the perforations and lids, so that liquid or vapour cannot leak out during heating.

5 In one embodiment, all plates and laminae have a series of holes drilled through (6) for locking means, which can be bolts which are used to assemble the multiautoclave and ensure a sufficient counter-pressure to prevent leakage when the liquid in the chambers is heated to high temperatures, and the bolts are placed in such a manner and their number is adjusted so that a sufficiently distributed even load is obtained in order to ensure that all
10 the chambers are tight when in use. The multiautoclave may alternatively be closed by pressing the plates against each other employing a clamping or squeezing mechanism that makes through-going bolts unnecessary. The squeezing mechanism may include springs or the like, which ensure the maintenance of a suitable pressure. A frame made of a rigid material that ensures good tightness in the outer chambers may enclose the entire
15 multiautoclave, also counteracting deformation of plates made of pure Teflon or another ductile material. Bolts and frames or whatever other locking mechanisms used are tightened with a torque wrench in order to ensure correct load throughout. A possible feature of the design is that a large number of plates may be placed on top of each other forming layers of reaction chambers according to the desired capacity. As an example, 10
20 plates with pressure chambers can be placed on top of each other without requiring a total multiautoclave thickness of more than typically 25 – 40 cm. Another important feature of this invention is that the large number of chambers which are created by the described assembly of the various described parts subsequent to performing e. g. a number of syntheses can be opened in both ends. This makes simple and efficient recovery and
25 washing of the synthesis products possible, and as an example on how this can be performed, the top plate with associated lid (e.g. septum) is removed first, and a filter paper placed over it and pressed against the plate with a sponge or another suitable absorbent or a filter supported by a rigid grid connected to a pump. The multiautoclave is then turned upside-down, and the bottom plate with associated lid is removed. The liquid
30 in the reaction chambers can then drain through the filter, and the synthesis product can be washed by flushing down through the perforations. As a result, the final, washed synthesis products are each in its own position on the filter paper, and these positions correspond to the positions of the perforations of the mainblock in the multiautoclave, so that identification of the synthesis products can be achieved. The samples are placed in a well-
35 defined matrix that, in principle, in a simple manner can be transferred to an automatic sample-switching unit for analysis, e.g. by X-ray diffraction. It is often desirable to calcine inorganic samples after synthesis, and for this purpose the samples can be washed

from the multiautoclave and into the perforations of another block which is made of a material suitable for performing calcination s.a. stainless steel or quartz.

Whenever the perforations, which on closing form the pressure chambers are going
5 through the central block, the bottom has to be closed before charging with liquid or other reactants. This can be achieved in a simple manner by placing the perforated plate (2) on top of a plate (7a) with balls, septa or other sealing devices between them. The two plates (2 and 7a) are then bolted together with a set of bolts (9) with lengths somewhat less than the combined thickness of the two plates, so that no part of the bolts is protruding from
10 the plate assembly. In the example shown in Figure 2, six bolts placed as illustrated have been found to be sufficient to hold the bottom plate sufficiently tight to the central block. After the chambers have been charged, the top plate (7b) with its sealing devices is put in place, and another set of bolts (11), which are considerably longer than the thickness of the entire multiautoclave, is put through a separate set of through-going holes (6) and
15 tightened with nuts (12) at the bottom side with a torque wrench and with a load sufficient to keep the multiautoclave tight under the prevailing synthesis conditions it is going to be exposed to. Springs adjusted to a suitable pressure can, for instance, be put on the bolts before placing the nuts on them.

20 The advantages of the present invention are primarily related to the large rationalization gain that gives a correspondingly large economic saving. The saving is estimated to be from 90 to 99%. In other words, either the cost related to a given synthesis program is reduced by 90 - 99%, or it is possible for a given amount of money to perform 10 - 100 times as many syntheses. Such an automated layout will make it possible to perform e.g.
25 1000 syntheses/formulations simultaneously, and it will thus be very useful for all research laboratories, in industry as well as in research institutions/ universities.

Multiautoclaves suited for the purposes mentioned above might be designed as described in the following examples, but the descriptions are to be considered merely as examples
30 of possible designs and the given measurements and other details shall not be considered to be limitations to the invention.

Example 1

A multiautoclave is built from 5 layers as shown in Figure 1. The different layers are
35 placed on top of each other and bolted together so that 100 hermetically sealed chambers (1) are formed. By stacking several layers in a suitable way, it is possible to make a multiautoclave with, e.g. 1000 chambers or more. A decisive element in the invention described here is the combination of steel balls (4) and a thin polymer film (3) for the

closing of the small chambers in the multiautoclave. In as much as the contact between the edge of the perforation and the steel ball is exposed to the total pressure developed by tightening the bolts, the multiautoclave will be tight aided by slight deformation of the ductile parts.

5

The multiautoclave is from bottom to top built up from the following elements as shown in Figure 1. Bottom plate (7a) made from aluminium or steel, with 100 symmetrically positioned cavities (8), each with a diameter of 13 mm. Each cavity has a depth of 8 mm, and stainless steel balls (4) with diameters of 13 mm are placed in each cavity. In addition, the plate has 9 smaller, through-going holes (6) for bolts that are used to keep all the plates bolted together. Furthermore, a thin polymer film (3a) made of Teflon, and the purpose of this film is to tighten against the perforations in the perforated plate (2) and to avoid direct contact between the steel balls (4) and the synthesis mixtures. The polymer film has 9 smaller holes (6) through it for bolts used to keep all the plates together. Further, a 2 cm thick Teflon plate (2) with 100 symmetrically positioned perforations with diameter 8 mm. In addition, the plate has 9 smaller holes (6) going through it for bolts used to keep all the plates together. There is also another set of holes (10) through the bottom plate (7a) and the perforated plate (2) that is used to keep these two plates with the corresponding balls tightly together while adding liquid. Furthermore, another thin polymer film (3b) made from 0.5 mm Teflon, and the purpose and design of this film is the same as for (3a). A top plate (7b) is placed above this, identical to the bottom plate (7a), but inverted in relation to it.

A frame ensuring good tightening of the outer chambers by preventing lateral deformation of the Teflon block (2) is fastened around the multiautoclave (not shown).

25

Example 2

In another embodiment, a multiautoclave was designed and built as shown in Figure 3. In this case, Teflon-lined septa (5) type "MICROSEP F138" from Alltech which are stable up to 250°C were used as bottom- and top-lids for each of the small chambers.

30

The multiautoclave was constructed from the following elements according to Figure 3. A bottom aluminium plate (7a), 2 cm thick with 9 smaller, through-going holes for bolts (11) used for holding all the plates bolted together. Above this plate a 2 cm thick Teflon plate with 100 symmetrically positioned perforations with diameters 8 mm is mounted. Further, above and below each perforation there is a 1 mm deep recess with diameter 13 mm where Teflon-lined septa (5) with thickness of 2 mm are placed. This plate also has 9 smaller, through-going holes for bolts used to keep all the plates bolted together. There is

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also a set of 6 holes (10) with associated bolts (9) used to hold the bottom plate (7a) and the central block (2) together.

5 An upper aluminium plate (7b), 2 cm thick, is placed on top. This plate also has 9 smaller, through-going holes for bolts (11) used to keep all the plates bolted together.

10 In order to test the designed multiautoclave, the bottom plate (7a) and the central block (2) (Figure 3) were bolted together so that a plate with 100 wells and solid bottom embedding the septa (5) was formed. The resulting were each filled with 0.5 ml water, and a top plate with the associated septa was fastened to it by bolts. The multiautoclave was then placed in a heating cabinet at 150°C for three days. When the multiautoclave was opened after three days, the liquid levels in the 100 chambers were unchanged.

Example 3

15 In yet another embodiment, a heavy duty multiautoclave was designed and built essentially as shown in Figure 3, but in this case the 20 mm thick central block (2) was made of 316-stainless steel with 13.75 mm wells which were 19 mm deep. The remaining 1 mm of the plate was drilled through with a diameter of 12.45 mm resulting in a design, essentially, as displayed in Figure 4. The 13.75 mm perforations were lined with Teflon
20 cylinders (13) with 2.75 mm wall thickness and 18 mm length so that above and below each cylinder there is a 1 mm deep recess with diameter 13.75 in one end and 12.45 in the other end. 3 mm thick Teflon disks with 13.75 mm and 12.45 mm diameters, respectively, (5) were used as bottom- and top lids for each of the small chambers.

25 Example 4

The bottom plate (7a) and the central block (2) of the multiautoclave described in Example 2 with septa (5) embedded were bolted together so that a plate with 100 wells with solid bottom was formed. This plate was then connected to an automatic pipetting machine of the "Tecan miniprepTM" type which was programmed to dispense the desired
30 amounts from 4 different solutions to each of the 100 wells, so that a total of 100 reaction mixtures with different compositions was prepared in about 30 min.

Example 5

35 A multiautoclave as described in Example 1 was used for 64 zeolite syntheses based on 56 different gel compositions derived from 4 different solutions, including water, so that it covered the composition range for the Na - Si - Al system shown in Figure 6. The matrix had 64 points, but 8 of these were excluded because they appeared outside the desired range. The 8 spare chambers were instead used for reproductions, so that the total

number of different compositions were 56, and these 56 compositions are given as molecular ratios in Table 1 and as volume units in Table 2, and the last 5 columns in Table 2 are volume units adjusted so that each small synthesis chamber will contain an exact volume of 0.5 ml synthesis mixture.

5

The four solutions were prepared so that all Si is dissolved in solution no. 4 and all Al is dissolved in solution no. 1. Solution no. 1 also contains an amount of NaOH sufficient to keep Al dissolved as well as water as solvent. Solution no. 3 was used to make-up the NaOH concentration (corrected for Na content in solutions no. 1 and no. 4), and finally, 10 water was used to make-up the amount of water required for the recipe. All solutions were normalized to 500 μ l. The compositions of the four solutions were:

1. 100 g NaAlO_2 + 8.5 g NaOH + 391.5 g water
2. 21.8 g NaOH + 100 g water
- 15 3. Water
4. Ludox LS-30 which is 30% silica sol in water

The solutions were prepared in the order and amounts given in Table 1, and automatic pipettes were used for charging the multiautoclave chambers with the solutions. Four gel 20 compositions were chosen as reproductions, and three parallels of these were made in the multiautoclave and, in addition, a larger scale (approx. 40 g gel) synthesis was performed in a separate plastic flask as a control of the possible effects of down- or up scaling. The four reproductions were chosen so that they represented points in the range of the four zeolitic phases FAU, CHA, GML and LTA given in Figure 4.5.c on page 70 in "Zeolite 25 Molecular Sieves, Structure Chemistry and Use" by D. W. Breck, 1974, Wiley and Sons, New York.

The multiautoclave was placed in a heating cabinet after being kept over-night at room temperature. It was opened after 93 hours of crystallization at 100°C, and it was 30 established that all 64 chambers had stayed tight without leakage, and a large filter paper was placed so that it covered the 64 perforations and a moisture absorbing material was pressed against the outside of the filter paper so that remaining moisture was separated from the crystalline products. The products were washed by adding a small aliquot of water (ca. 0.5 ml) to each perforation for washing. Thereafter, when the solid material was 35 transferred to the filter paper, the perforated Teflon plate was removed and the 64 filter "cakes" were transferred to 64 separate glass sample vials. The samples were then dried at 100°C in a drying cabinet.

TABLE 1

SiO2/Al2O3	Na2O/Al2O3	SiO2/Na2O	sum	SiO2	Na2O	Al2O3	H2O
1.0	1.5	0.654	3.53	0.283	0.433	0.283	19.0
1.0	1.9	0.526	3.90	0.256	0.487	0.256	19.0
1.0	2.4	0.417	4.40	0.227	0.545	0.227	19.0
1.0	3.0	0.333	5.00	0.200	0.600	0.200	19.0
1.0	4.0	0.250	6.00	0.167	0.667	0.167	19.0
1.5	1.5	0.980	4.03	0.372	0.380	0.248	19.0
1.5	1.9	0.789	4.40	0.341	0.432	0.227	19.0
1.5	2.4	0.625	4.90	0.306	0.490	0.204	19.0
1.5	3.0	0.500	5.50	0.273	0.545	0.182	19.0
1.5	4.0	0.375	6.50	0.231	0.615	0.154	19.0
1.5	5.3	0.283	7.80	0.192	0.679	0.128	19.0
1.5	8.0	0.188	10.50	0.143	0.762	0.095	19.0
2.3	1.5	1.503	4.83	0.476	0.317	0.207	19.0
2.3	1.9	1.211	5.20	0.442	0.365	0.192	19.0
2.3	2.4	0.958	5.70	0.404	0.421	0.175	19.0
2.3	3.0	0.767	6.30	0.365	0.476	0.159	19.0
2.3	4.0	0.575	7.30	0.315	0.548	0.137	19.0
2.3	5.3	0.434	8.60	0.267	0.616	0.116	19.0
2.3	8.0	0.288	11.30	0.204	0.708	0.088	19.0
2.8	1.5	1.830	5.33	0.525	0.287	0.188	19.0
2.8	1.9	1.474	5.70	0.491	0.333	0.175	19.0
2.8	2.4	1.167	6.20	0.452	0.387	0.161	19.0
2.8	3.0	0.933	6.80	0.412	0.441	0.147	19.0
2.8	4.0	0.700	7.80	0.359	0.513	0.128	19.0
2.8	5.3	0.528	9.10	0.308	0.582	0.110	19.0
2.8	8.0	0.350	11.80	0.237	0.678	0.085	19.0
4.0	1.5	2.614	6.53	0.613	0.234	0.153	19.0
4.0	1.9	2.105	6.90	0.580	0.275	0.145	19.0
4.0	2.4	1.667	7.40	0.541	0.324	0.135	19.0
4.0	3.0	1.333	8.00	0.500	0.375	0.125	19.0
4.0	4.0	1.000	9.00	0.444	0.444	0.111	19.0
4.0	5.3	0.755	10.30	0.388	0.515	0.097	19.0
4.0	8.0	0.500	13.00	0.308	0.615	0.077	19.0
4.0	12.0	0.333	17.00	0.235	0.706	0.059	19.0
6.0	1.5	3.922	8.53	0.703	0.179	0.117	19.0
6.0	1.9	3.158	8.90	0.674	0.213	0.112	19.0
6.0	2.4	2.500	9.40	0.638	0.255	0.106	19.0
6.0	3.0	2.000	10.00	0.600	0.300	0.100	19.0
6.0	4.0	1.500	11.00	0.545	0.364	0.091	19.0
6.0	5.3	1.132	12.30	0.488	0.431	0.081	19.0
6.0	8.0	0.750	15.00	0.400	0.533	0.067	19.0
6.0	12.0	0.500	19.00	0.316	0.632	0.053	19.0
10.0	1.5	6.536	12.53	0.798	0.122	0.080	19.0
10.0	1.9	5.263	12.90	0.775	0.147	0.078	19.0
10.0	2.4	4.167	13.40	0.746	0.179	0.075	19.0
10.0	3.0	3.333	14.00	0.714	0.214	0.071	19.0
10.0	4.0	2.500	15.00	0.667	0.267	0.067	19.0
10.0	5.3	1.887	16.30	0.613	0.325	0.061	19.0
10.0	8.0	1.250	19.00	0.526	0.421	0.053	19.0
10.0	12.0	0.833	23.00	0.435	0.522	0.043	19.0
20.0	1.5	13.072	22.53	0.888	0.068	0.044	19.0
20.0	2.4	8.333	23.40	0.855	0.103	0.043	19.0
20.0	4.0	5.000	25.00	0.800	0.160	0.040	19.0
20.0	5.3	3.774	26.30	0.760	0.202	0.038	19.0
20.0	8.0	2.500	29.00	0.690	0.276	0.034	19.0
20.0	12.0	1.667	33.00	0.606	0.364	0.030	19.0

TABLE 2

volume units of solution						microliter solution				
ludox LS-30	soln 1	soln 2	vann	sum	factor	ludox LS-30	soln 1	soln 2	vann	sum
46.82415	221.8	8.8032	80.2	357.68	1.398	65.45573	310.122	12.306	112.117	500
42.38186	200.8	44.782	67.85	355.81	1.405	59.55614	282.17	62.928	95.3453	500
37.56574	178	83.788	54.46	353.79	1.413	53.08987	251.534	118.41	76.963	500
33.05785	166.6	120.3	41.92	351.9	1.421	46.97017	222.539	170.92	59.566	500
27.54821	130.5	164.92	26.6	349.59	1.43	39.40061	186.676	235.88	38.0473	500
61.52206	194.3	7.5557	95.47	358.87	1.393	85.71586	270.741	10.527	133.016	500
56.34861	178	39.551	83.24	357.12	1.4	78.89315	249.191	55.374	116.541	500
50.59875	159.8	75.11	69.64	355.17	1.408	71.23129	224.99	105.74	98.0404	500
45.07889	142.4	109.25	56.59	353.3	1.415	63.79643	201.507	154.61	80.0876	500
38.14367	120.5	152.14	40.19	350.95	1.425	54.34291	171.647	216.75	57.2603	500
31.7864	100.4	191.45	25.16	348.8	1.433	45.56532	143.922	274.45	36.0653	500
23.61275	74.58	242	5.832	346.03	1.445	34.11934	107.769	349.68	8.42692	500
78.70917	162.1	6.0969	113.3	360.27	1.388	109.2369	225.022	8.4616	157.279	500
73.10871	150.6	33.273	101.7	358.69	1.394	101.9118	209.933	46.382	141.773	500
66.69566	137.4	64.393	88.4	356.87	1.401	93.4441	192.49	90.218	123.848	500
60.3437	124.3	95.216	75.22	355.08	1.408	84.9719	175.038	134.08	105.913	500
52.07744	107.3	135.33	58.06	352.75	1.417	73.81736	152.06	191.82	82.3009	500
44.20527	91.06	173.53	41.73	350.52	1.426	63.05645	129.893	247.53	59.5215	500
33.64295	69.3	224.78	19.81	347.54	1.439	48.40187	99.7054	323.39	28.4998	500
86.83113	148.9	5.4075	121.8	360.93	1.385	120.2887	203.541	7.4911	168.68	500
81.19472	137.4	30.245	110.6	359.44	1.391	112.9455	191.115	42.072	153.867	500
74.64676	126.3	59.099	97.66	357.72	1.398	104.3381	176.551	82.606	136.505	500
68.06028	115.2	88.123	84.63	355.98	1.405	95.59593	161.758	123.78	118.871	500
59.3346	100.4	128.57	67.37	353.68	1.414	83.88217	141.937	178.94	95.2423	500
50.85823	86.06	163.93	50.6	351.44	1.423	72.35622	122.434	233.22	71.9928	500
39.22118	66.37	215.2	27.58	348.38	1.435	56.29159	95.2511	308.87	39.5881	500
101.2492	119.9	4.1838	136.7	362.1	1.381	139.8085	165.599	5.7771	188.815	500
95.81986	113.5	24.767	126.7	360.81	1.386	132.7847	157.28	34.322	175.614	500
89.34554	105.8	49.312	114.8	359.27	1.392	124.3431	147.281	68.629	159.748	500
82.64463	97.89	74.717	102.4	357.68	1.398	115.5296	136.841	104.45	143.182	500
73.46189	87.01	109.53	85.49	355.49	1.406	103.3234	122.384	154.05	120.24	500
64.19	76.03	144.68	68.39	353.29	1.415	90.84579	107.604	204.76	96.788	500
50.85823	60.24	195.22	43.8	350.12	1.428	72.62926	86.0273	278.79	62.5493	500
38.89159	46.07	240.59	21.73	347.28	1.44	55.99491	66.3244	346.4	31.2843	500
116.2644	91.81	2.9093	152.3	363.32	1.376	160.0031	126.346	4.0038	209.647	500
111.431	87.99	18.92	143.9	362.27	1.38	153.7963	121.445	26.114	198.645	500
105.5038	83.31	38.554	133.6	360.98	1.385	146.1356	115.396	53.402	185.067	500
99.17355	78.31	59.523	122.6	359.6	1.39	137.8934	108.887	82.762	170.457	500
90.15778	71.19	89.388	106.9	357.64	1.398	126.0449	99.531	124.97	149.456	500
80.62891	63.67	120.95	90.32	355.57	1.406	113.38	89.5303	170.08	127.007	500
66.1157	52.21	169.03	65.06	352.41	1.419	93.80433	74.0723	239.81	92.309	500
52.19661	41.22	215.13	40.84	349.39	1.431	74.69769	58.9848	307.88	58.4422	500
131.9148	62.5	1.581	168.6	364.59	1.371	180.9081	85.7122	2.1682	231.212	500
128.1312	60.71	12.665	162.3	363.83	1.374	176.0871	83.428	17.406	223.079	500
123.3502	58.44	26.672	154.4	362.87	1.378	169.9663	80.5281	36.751	212.754	500
118.0638	55.94	42.159	145.6	361.8	1.382	163.1606	77.3036	58.262	201.274	500
110.1928	52.21	65.217	132.6	360.22	1.388	152.9531	72.4674	90.525	184.055	500
101.4045	48.04	90.964	118	358.45	1.395	141.4491	67.017	126.89	164.649	500
86.99435	41.22	133.18	94.16	355.55	1.406	122.3386	57.9626	187.29	132.411	500
71.86489	34.05	177.5	69.09	352.5	1.418	101.9356	48.2959	251.77	97.9937	500
146.7281	34.76	0.3237	184	365.79	1.367	200.5611	47.5118	0.4424	251.485	500
141.2729	33.47	14.739	175.3	364.76	1.371	193.6507	45.8747	20.203	240.271	500
132.2314	31.32	38.63	160.9	363.05	1.377	182.1111	43.1411	53.201	221.546	500
125.6953	29.78	55.901	150.4	361.81	1.382	173.7011	41.1488	77.25	207.9	500
113.9926	27	86.824	131.8	359.6	1.39	158.4988	37.5475	120.72	183.231	500
100.1753	23.73	123.33	109.7	356.99	1.401	140.3068	33.2379	172.74	153.712	500

R = point reproduced

Example 6

In another embodiment, pre-mixed gels of aluminumphosphate, silicoaluminumphosphate and cobaltaluminumphosphate were prepared and administered by means of the "Tecan MiniprepTM" to the 100 cavities of the multiautoclave of example 3. Thereafter 6 different organic compounds were added according to the molar ratios specified in Table 3. The multiautoclave was then closed and was heated for 48 h in an oven at 200°C. The solid synthesis products were recovered as described in example 5 and were dried overnight at ambient temperatures. The products were characterised by X-ray diffraction using a Siemens D-5000 diffractometer equipped with PSD detector and an automatic sample shifter. Several different crystalline phases were identified among the products including those of the structures AFI, ERI and CHA.

TABLE 3

Position	Mols AlPO gel	Mols SAPO gel	Mols CoAPO gel	R1	Mols R1	R2	Mols R2
1	1	0	0	CYCLOHEXY	1	-	0
2	1	0	0	MORPHOLIN	1	-	0
3	1	0	0	TRIPROPYL	1	-	0
4	1	0	0	DI-N-PROP	1	-	0
5	1	0	0	TEAOH	1	-	0
6	1	0	0	TRIETHYLA	1	-	0
7	0	1	0	CYCLOHEXY	1	-	0
8	0.25	0.75	0	CYCLOHEXY	1	-	0
9	0.5	0.5	0	CYCLOHEXY	1	-	0
10	0	1	0	MORPHOLIN	1	-	0
11	0.25	0.75	0	MORPHOLIN	1	-	0
12	0.5	0.5	0	MORPHOLIN	1	-	0
13	0	1	0	TRIPROPYL	1	-	0
14	0.25	0.75	0	TRIPROPYL	1	-	0
15	0.5	0.5	0	TRIPROPYL	1	-	0
16	0	1	0	DI-N-PROP	1	-	0
17	0.25	0.75	0	DI-N-PROP	1	-	0
18	0.5	0.5	0	DI-N-PROP	1	-	0
19	0	1	0	TEAOH	1	-	0
20	0.25	0.75	0	TEAOH	1	-	0
21	0.5	0.5	0	TEAOH	1	-	0
22	0	1	0	TRIETHYLA	1	-	0
23	0.25	0.75	0	TRIETHYLA	1	-	0
24	0.5	0.5	0	TRIETHYLA	1	-	0
25	0	0	1	CYCLOHEXY	1	-	0
26	0.25	0	0.75	CYCLOHEXY	1	-	0
27	0.5	0	0.5	CYCLOHEXY	1	-	0
28	0	0	1	MORPHOLIN	1	-	0
29	0.25	0	0.75	MORPHOLIN	1	-	0
30	0.5	0	0.5	MORPHOLIN	1	-	0
31	0	0	1	TRIPROPYL	1	-	0
32	0.25	0	0.75	TRIPROPYL	1	-	0
33	0.5	0	0.5	TRIPROPYL	1	-	0
34	0	0	1	DI-N-PROP	1	-	0
35	0.25	0	0.75	DI-N-PROP	1	-	0
36	0.5	0	0.5	DI-N-PROP	1	-	0
37	0	0	1	TEAOH	1	-	0
38	0.25	0	0.75	TEAOH	1	-	0
39	0.5	0	0.5	TEAOH	1	-	0
40	0	0	1	TRIETHYLA	1	-	0
41	0.25	0	0.75	TRIETHYLA	1	-	0
42	0.5	0	0.5	TRIETHYLA	1	-	0
43	0	0.25	0.75	CYCLOHEXY	1	-	0
44	0	0.5	0.5	CYCLOHEXY	1	-	0
45	0	0.75	0.25	CYCLOHEXY	1	-	0
46	0	0.25	0.75	MORPHOLIN	1	-	0
47	0	0.5	0.5	MORPHOLIN	1	-	0
48	0	0.75	0.25	MORPHOLIN	1	-	0
49	0	0.25	0.75	TRIPROPYL	1	-	0

Table 3, gel compositions for MeAPO syntheses, continued.

Position	Mols AIPO gel	Mols SAPO gel	Mols CoAPO gel	R1	Mols R1	R2	Mols R2
50	0	0.5	0.5	TRIPROPYL	1	-	0
51	0	0.75	0.25	TRIPROPYL	1	-	0
52	0	0.25	0.75	DI-N-PROP	1	-	0
53	0	0.5	0.5	DI-N-PROP	1	-	0
54	0	0.75	0.25	DI-N-PROP	1	-	0
55	0	0.25	0.75	TEAOH	1	-	0
56	0	0.5	0.5	TEAOH	1	-	0
57	0	0.75	0.25	TEAOH	1	-	0
58	0	0.25	0.75	TRIETHYLA	1	-	0
59	0	0.5	0.5	TRIETHYLA	1	-	0
60	0	0.75	0.25	TRIETHYLA	1	-	0
61	0	0	1	CYCLOHEXY	0.25	MORPHOLIN	0.75
62	0	0	1	CYCLOHEXY	0.25	TRIPROPYL	0.75
63	0	0	1	CYCLOHEXY	0.25	DI-N-PROP	0.75
64	0	0	1	CYCLOHEXY	0.25	TEAOH	0.75
65	0	0	1	CYCLOHEXY	0.25	TRIETHYLA	0.75
66	0	0	1	MORPHOLIN	0.25	TRIPROPYL	0.75
67	0	0	1	MORPHOLIN	0.25	DI-N-PROP	0.75
68	0	0	1	MORPHOLIN	0.25	TEAOH	0.75
69	0	0	1	MORPHOLIN	0.25	TRIETHYLA	0.75
70	0	0	1	TRIPROPYL	0.25	DI-N-PROP	0.75
71	0	0	1	TRIPROPYL	0.25	TEAOH	0.75
72	0	0	1	TRIPROPYL	0.25	TRIETHYLA	0.75
73	0	0	1	DI-N-PROP	0.25	TEAOH	0.75
74	0	0	1	DI-N-PROP	0.25	TRIETHYLA	0.75
75	0	0	1	TEAOH	0.25	TRIETHYLA	0.75
76	0	0	1	CYCLOHEXY	0.75	MORPHOLIN	0.25
77	0	0	1	CYCLOHEXY	0.75	TRIPROPYL	0.25
78	0	0	1	CYCLOHEXY	0.75	DI-N-PROP	0.25
79	0	0	1	CYCLOHEXY	0.75	TEAOH	0.25
80	0	0	1	CYCLOHEXY	0.75	TRIETHYLA	0.25
81	0	0	1	MORPHOLIN	0.75	TRIPROPYL	0.25
82	0	0	1	MORPHOLIN	0.75	DI-N-PROP	0.25
83	0	0	1	MORPHOLIN	0.75	TEAOH	0.25
84	0	0	1	MORPHOLIN	0.75	TRIETHYLA	0.25
85	0	0	1	TRIPROPYL	0.75	DI-N-PROP	0.25
86	0	0	1	TRIPROPYL	0.75	TEAOH	0.25
87	0	0	1	TRIPROPYL	0.75	TRIETHYLA	0.25
88	0	0	1	DI-N-PROP	0.75	TEAOH	0.25
89	0	0	1	DI-N-PROP	0.75	TRIETHYLA	0.25
90	0	0	1	TEAOH	0.75	TRIETHYLA	0.25

91-100

Reproductions chosen at random from compositions 1 - 90

CYCLOHEX = Cyclohexylamine

MORPHOLI = morpholine

5 TRIPROPYL = tripropylamine

DI-N-PROP = dipropylamine

TEAOH = tetraethylammonium hydroxide

TRIETHYLA = triethylamine

PATENT CLAIMS

1. A pressure and temperature reactor vessel comprising
 - a) a central block having a multitude of perforations, wherein said perforations are through-going perforations, or cavities or other form of holes permanently closed at one end,
 - b) cover means, operatively associated with a sealing means, for engagement with said central block to seal the open ends of said perforations forming a multitude of chambers,
 - c) a sealing means, operatively associated with the covers means, to form a pressure tight seal when said cover means is brought into position by a locking means,
 - d) a locking means acting in concert with the cover means to engage the sealing means so as to define a multitude of reaction chambers.
2. A pressure and temperature reactor vessel as claimed in claim 1, wherein said perforations are through-going perforations.
3. A pressure and temperature reactor vessel as claimed in claim 1, wherein said sealing means is wholly or partly constructed of a compressible or deformable material.
4. A pressure and temperature reactor vessel as claimed in claim 1, wherein said central block is wholly or partly constructed of a compressible or deformable material.
5. A pressure and temperature reactor vessel as claimed in claim 1, wherein said sealing means is permanently fixed to the cover means.
6. A pressure and temperature reactor vessel as claimed in claim 1, wherein said cover means, acting in concert with the sealing means, has a multitude of protrusions of spherical shape, form or profile, either in part or wholly, presenting a curved surface towards the openings of the multitude of perforations in the central block.
7. A pressure and temperature reactor vessel as claimed in claim 1, wherein the sealing means consists of spherical shaped bodies or similar shaped components, permanently fixed to the cover means, such that they present a curved surface towards the opening of the multitude of perforations in the central block.
8. A pressure and temperature reactor vessel as claimed in claim 1, wherein the sealing means consists of spherical shaped bodies or similar shaped components, loosely fixed to

the cover means, such that they present a curved surface towards the opening of the multitude of perforations in the central block.

9. A pressure and temperature reactor vessel as claimed in claim 1, wherein the sealing means consists of circular disks or other similar circular-shaped components that may be permanently fastened to the cover means.

10. A pressure and temperature reactor vessel as claimed in claim 1, wherein the sealing means consists of circular disks or other similar circular-shaped components that may be loosely fastened to the cover means.

11. A pressure and temperature reactor vessel as claimed in claim 1, wherein the sealing means consists of a flat plate or membrane, covering all the chambers, that may be permanently fastened to the cover means.

12. A pressure and temperature reactor vessel as claimed in claim 1, wherein the sealing means consists of a flat plate or membrane, covering all the chambers, that may be loosely fastened to the cover means.

13. A pressure and temperature reactor vessel as claimed in claim 1, wherein said locking means acts in concert with a single cover means on one surface of the central block to define a multitude of cavities.

14. A pressure and temperature reactor vessel as claimed in claim 1, wherein said locking means consists of a plurality of fasteners passing by means of appropriate holes from the upper surface of the upper cover means through the central block through to the lower surface of the lower cover means.

15. A pressure and temperature reactor vessel as claimed in claim 1, wherein said cover means is a rigid flat plate.

16. A pressure and temperature reactor vessel as claimed in claim 1, wherein said cover means, acting in concert with the sealing means, has a multitude of protruding profiles of dimensions sufficiently larger than the dimensions of the perforations in the central block and of cross sectional shape such that in concert with the sealing means through the action of the locking means, a pressure tight seal is formed over and around each of the multitude of perforations in the central block.

17. A pressure and temperature reactor vessel as claimed in claim 1, wherein said central block, acting in concert with the sealing means, has a multitude of protruding profiles of dimensions sufficiently larger than the dimensions of the perforations and of cross sectional shape such that in concert with the sealing means and through the action of the locking means on the cover means, a pressure tight seal is formed over and around each of the multitude of perforations in the central block.

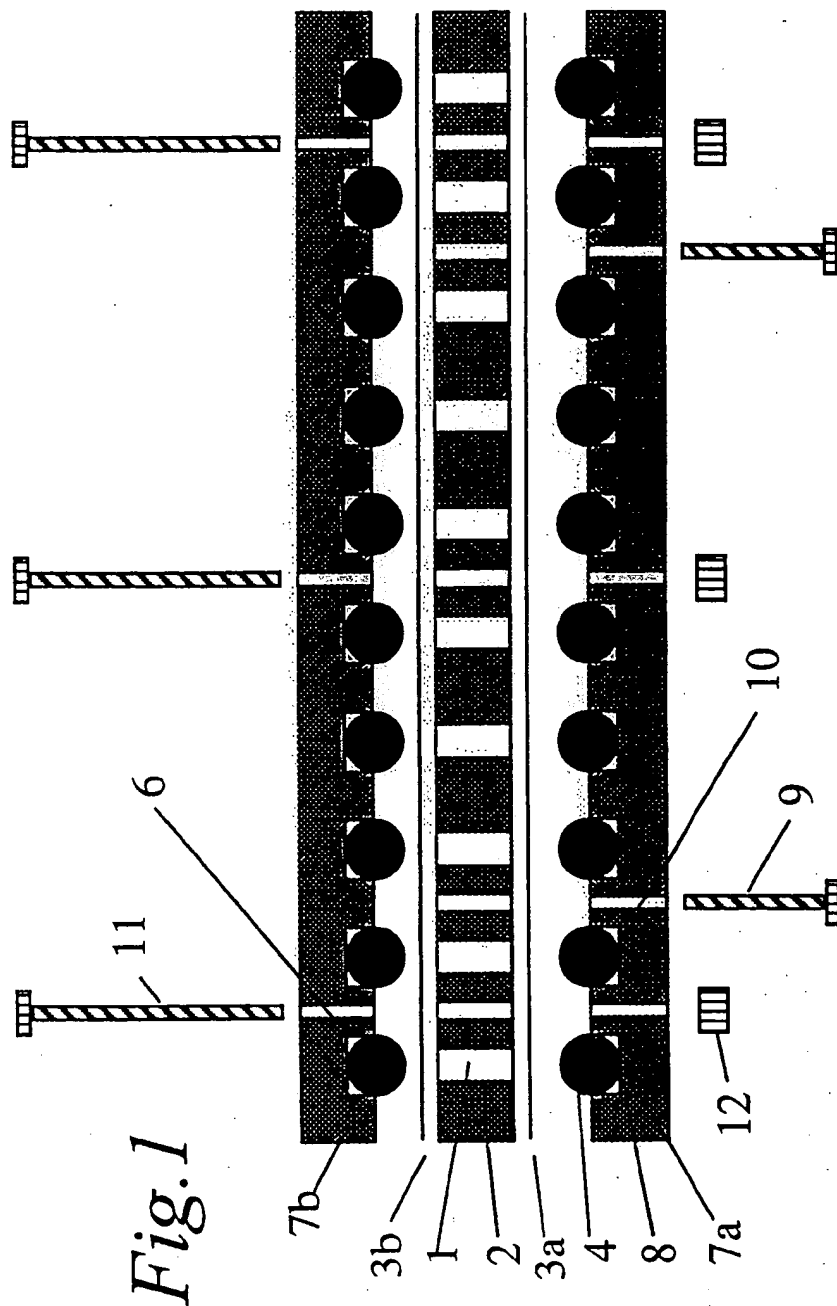


Fig. 1

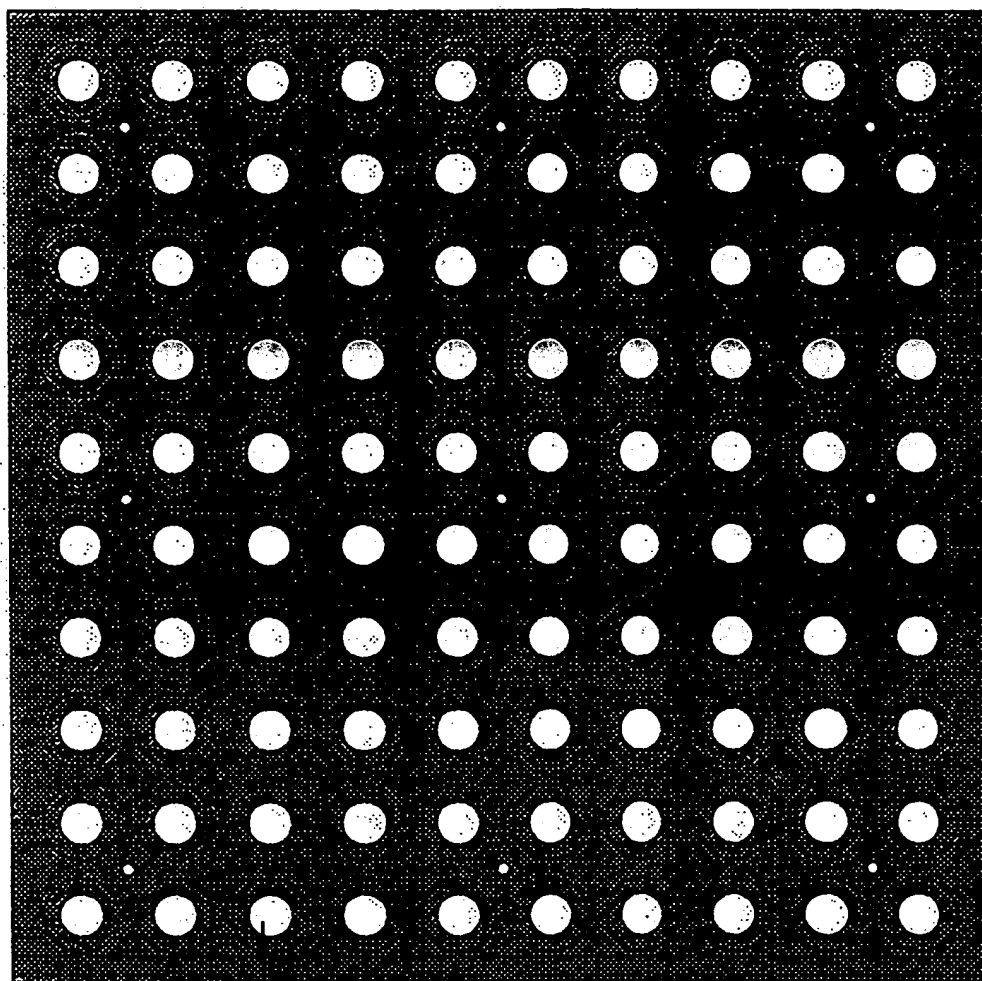


Fig. 2

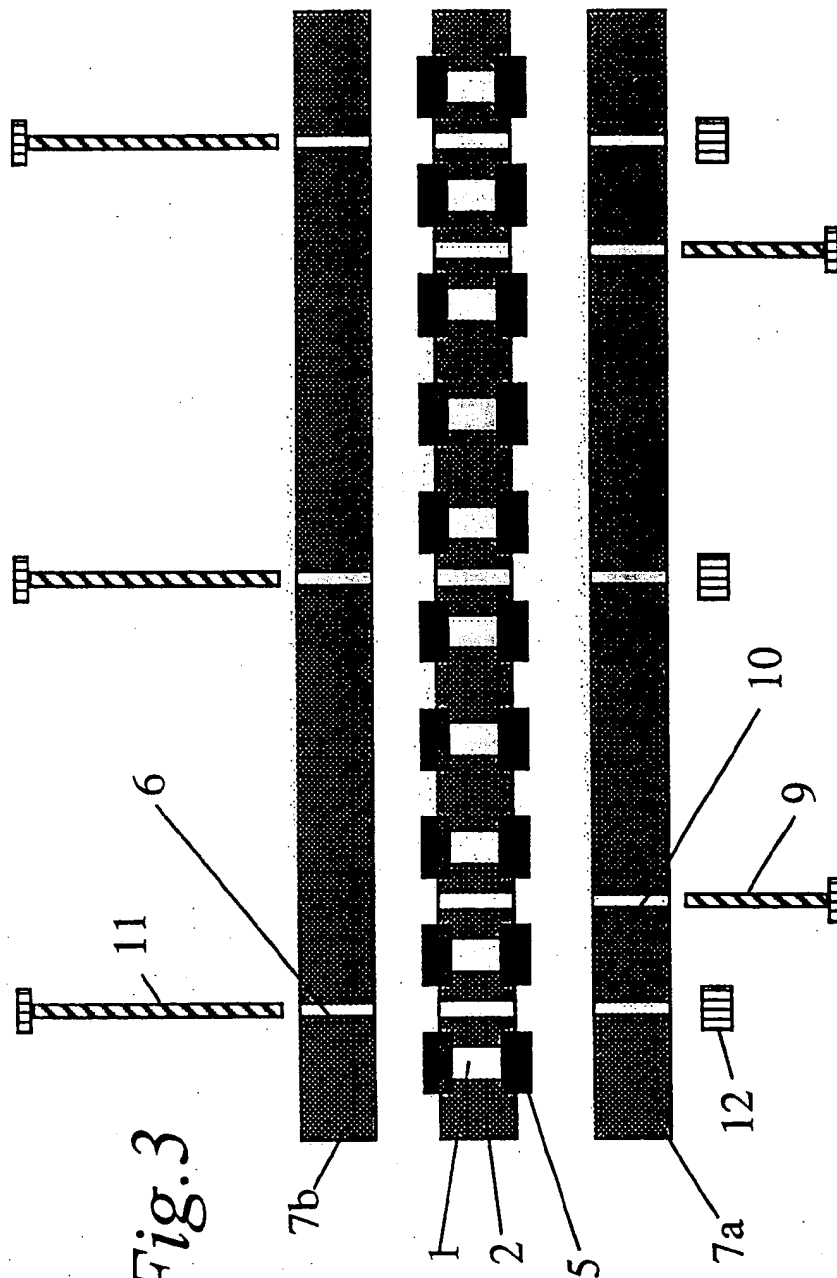


Fig.3

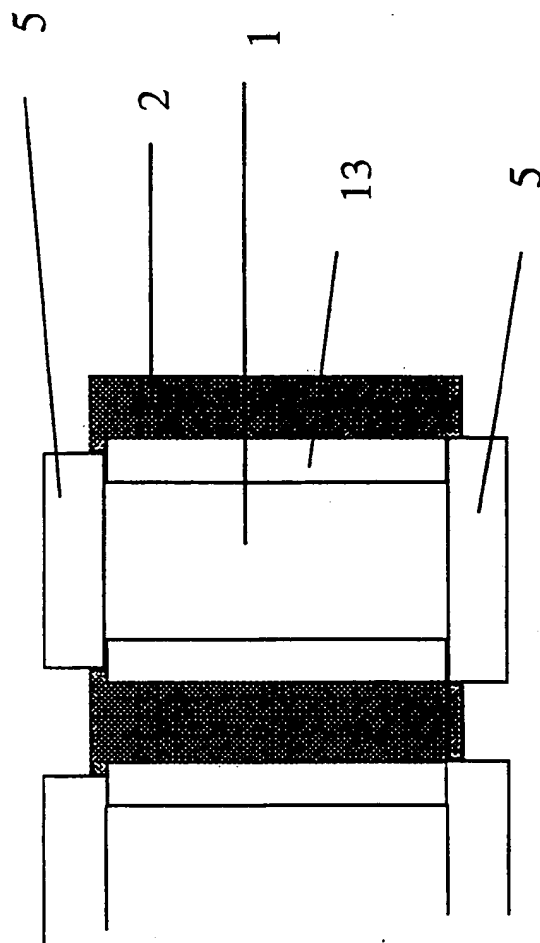
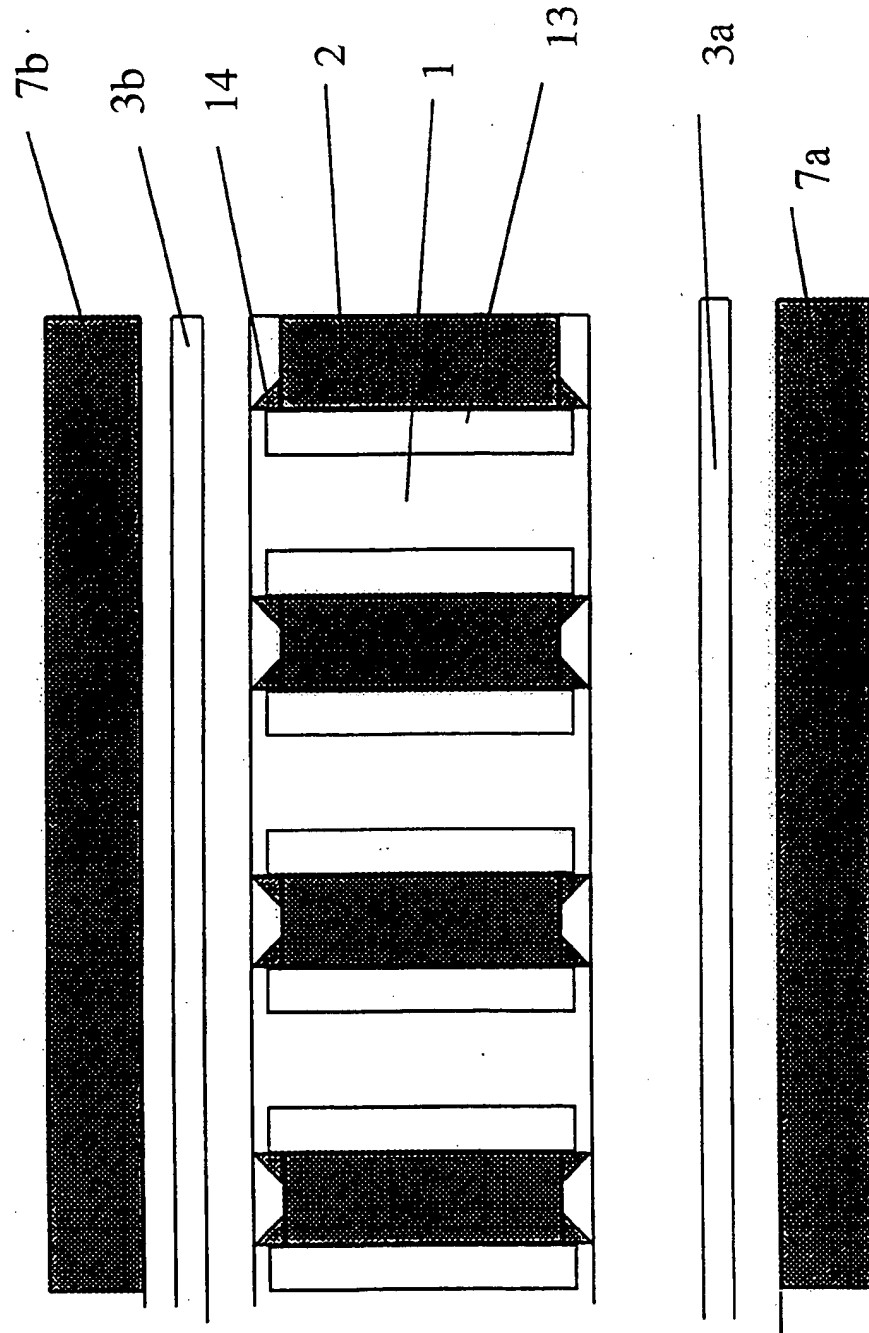


Fig. 4

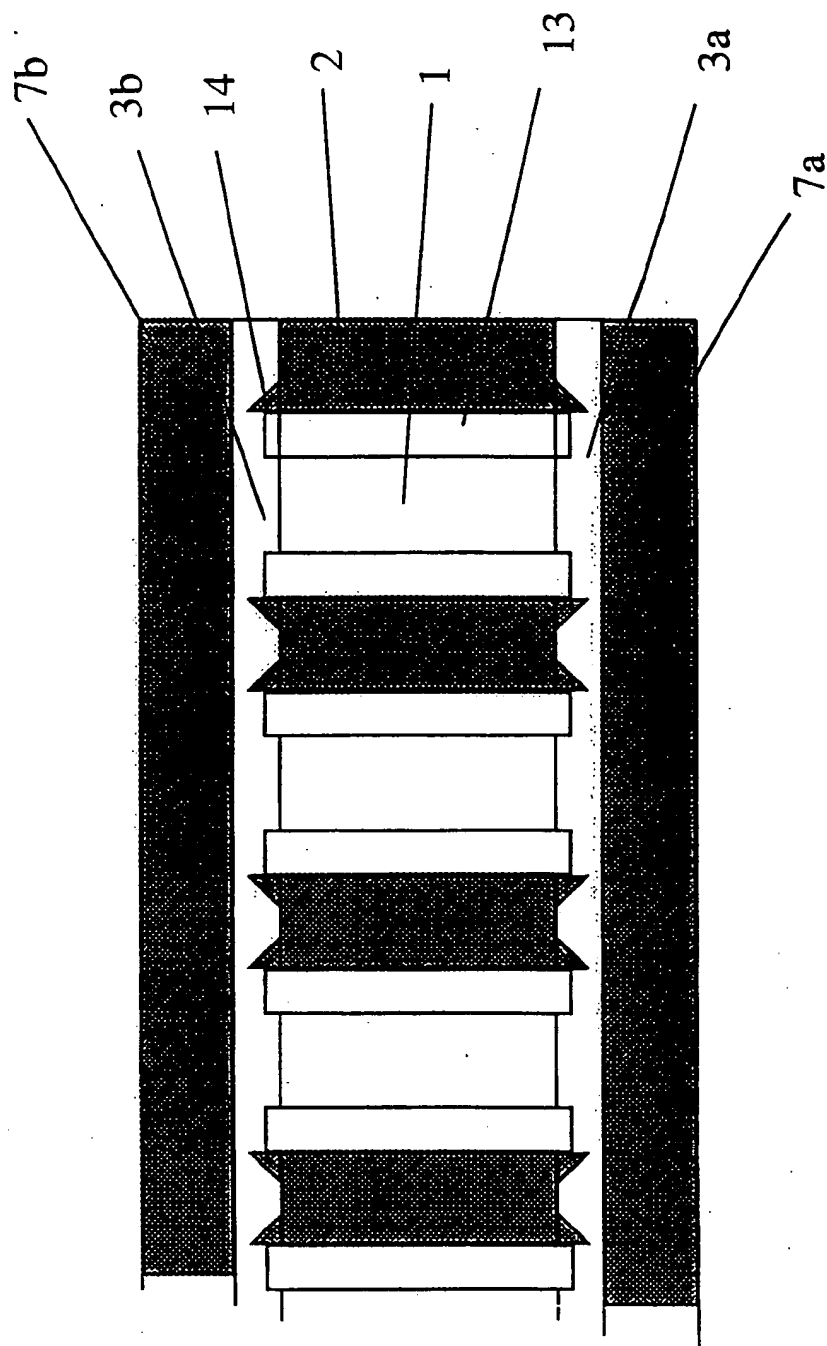
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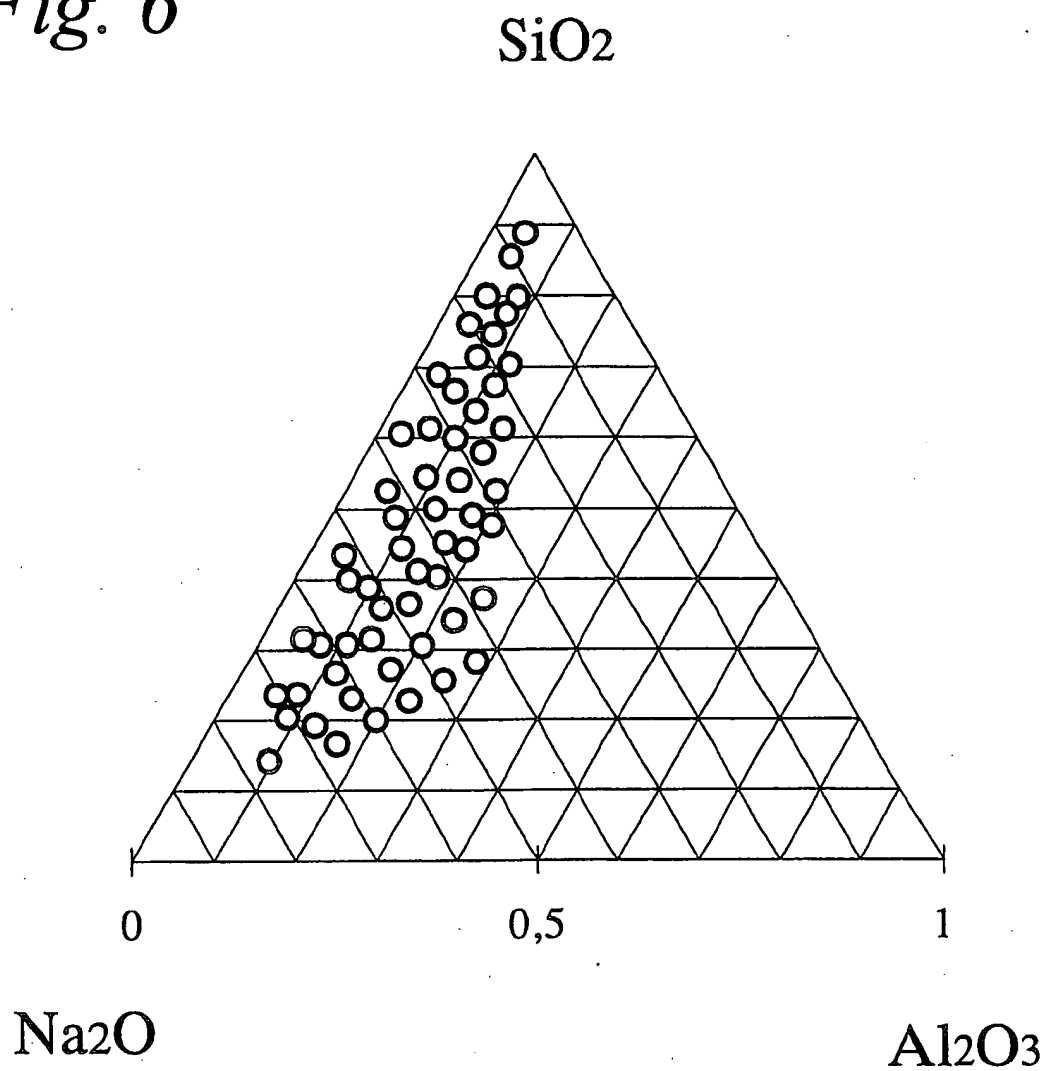
Fig 5a



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Fig. 5b





INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 98/00051

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: B01J 3/00, C30B 29/34

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: B01J, C01B; C30B, F17C, F16J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5112574 A (NIKKI HORTON), 12 May 1992 (12.05.92), column 3, line 65 - column 4, line 4, figures 1-4,9, abstract --	1-17
Y	US 5505916 A (BERNIE B. BERRY, JR.), 9 April 1996 (09.04.96), column 6, line 3 - line 7, figure 1 --	1-17
A	US 5400741 A (GEORGE T. DETITTA ET AL), 28 March 1995 (28.03.95), figures 1-7 --	1-17
A	US 5531185 A (KOJI ASANO ET AL), 2 July 1996 (02.07.96), figures 8-9, abstract --	1-12

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 98/00051

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	EP 0553539 A1 (SCHERING CORPORATION), 4 August 1993 (04.08.93), figures 1-7 --	1-17
P,A	EP 0780496 A1 (DORNIER GMBH), 25 June 1997 (25.06.97), figure 3 -- -----	1-17

INTERNATIONAL SEARCH REPORT
Information on patent family members

09/06/98

International application No.
PCT/NO 98/00051

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